## Estimation of the emission sources based on the relationship between atmospheric O2 and CO2 variations at Hateruma and Ochi-ishi

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Short-term variations in atmospheric  $CO_2$  concentration are usually associated with the inversely correlated changes in the  $O_2$  concentration. This is because the fact that the exchange between  $CO_2$  and  $O_2$  occurs during most source processes including fossil fuel burning and land biotic respiration/photosynthesis. The exchange ratios of  $O_2$  to  $CO_2$  depend on the elemental compositions of the organic materials produced or consumed during the individual source processes. For example, the exchange ratio is -1.1 for the land biotic processes, -1.95 for natural gas burning, -1.44 for gasoline burning, and -1.17 for coal burning. Therefore, slopes of the correlation plots between  $O_2$  and  $CO_2$  concentrations during the short-term event ( $D_-O_2/D_-CO_2$  ratio) could give some information on the  $CO_2$  sources process.

In-situ measurements of atmospheric  $O_2$  and  $CO_2$  concentrations have been conducted at Hateruma Island (HAT, Lat. 24.3N, Long. 123.49E) and Cape Ochi-ishi (COI, Lat. 43.1N, Long. 145.3E). These data often showed large variations with the synoptic-scale durations and there are inverse correlations between  $O_2$  and  $CO_2$  variations. The short-term events have been extracted from the time series by visual inspection, and the  $D_-O_2/D_-CO_2$  ratios for the events have been calculated. Based on backward trajectory calculation, we have classified the event by the source countries (China, Japan, Korea, and Taiwan) that mostly influenced the events.

Using the data observed at Hateruma during the period from October 2006 to August 2008, we have found 50 events, including 22 China, 12 Japan, and 9 Korea events. The average  $D_{-}O_{2}/D_{-}CO_{2}$  ratios for China, Korea, and Japan events are -1.20+/-0.02, -1.40+/-0.05, and -1.45+/-0.06, respectively. Because these events occurred during late fall-early spring (November-April), the contribution of the biological emissions to the observed  $O_{2}$  and  $CO_{2}$  variations are relatively small. Therefore, the resulted  $D_{-}O_{2}/D_{-}CO_{2}$  ratios suggest that the fractional contribution of coal consumption to the total fossil fuel consumption is larger for China than for Korea and Japan. Actually, the  $O_{2}:CO_{2}$  exchange ratios for the fossil fuel emissions from the individual countries, calculated from national fossil fuel emission inventories from CDIA data, show good agreements with the observed  $D_{-}O_{2}/D_{-}CO_{2}$  ratios. On the other hand, the average of  $D_{-}O_{2}/D_{-}CO_{2}$  ratios for the 76 short-term events in the observations at COI (March 2005-December 2007) is -1.14+/-0.03. Because most of the events occurred during summer-early fall (June-October), the biological emissions probably contributed to the observed  $CO_{2}$  and  $O_{2}$  variations at COI.